High temperature series expansion study of the Heisenberg antiferromagnet on the hyperkagome lattice: Comparison with $Na_4Ir_3O_8$.

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We develop high temperature series expansions for $\ln Z$ and the uniform structure factor of the spin-half Heisenberg model on the hyperkagome lattice to order β^{16}. These expansions are used to calculate the uniform susceptibility (χ), the entropy (S), and the heat capacity (C) of the model as a function of temperature. Series extrapolations of the expansions converge well down to a temperature of approximately $J/4$. A comparison with the experimental data for $Na_4Ir_3O_8$ shows that its magnetic susceptibility is reasonably well described by the model with an exchange constant $J \approx 300K$, but there are also additional smaller terms present in the system. The specific heat of the model has two peaks. The lower temperature peak, which is just below our range of convergence contains about 40 percent of the total entropy. Despite being a 3-dimensional lattice, this model shares many features with the kagome lattice Heisenberg model and the material must be considered a strong candidate for a quantum spin-liquid.

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INTRODUCTION

Despite many experimental and theoretical studies, the spin-half kagome lattice Heisenberg Model remains incompletely understood and a subject of intense ongoing research.[1–3] The kagome lattice is a two-dimensional lattice of corner sharing triangles, where classical antiferromagnetic models lead to exponential ground state degeneracy. Whether quantum fluctuations lift this degeneracy and lead to an ordered state, or select a valence-bond order, or lead to a resonating valence-bond quantum spin-liquid possibly with topological order and fractional excitations remains a topic of great interest. Numerical evidence shows that many distinct singlet states are very close to each other in energy.[4–13]

The hyperkagome lattice is a three-dimensional lattice of corner sharing triangles, where similar ground state degeneracy at the classical level and possible competing phases with nearly the same energy in presence of strong quantum fluctuations remains valid. Experimental discovery[14] of the material $Na_4Ir_3O_8$, where the Ir ions form a hyperkagome lattice, with strong signatures of a gapless quantum spin-liquid phase has led to much excitement. Theoretical proposals for this system also include various quantum spin-liquids as well as valence bond order.[15–18]

In this paper, we develop high temperature series expansions for $\ln Z$ and the uniform structure factor of the spin-half hyperkagome lattice Heisenberg model to order β^{16}. These series expansions are used to calculate the uniform susceptibility, the entropy and the heat capacity of the model. Padé and d-log Padé approximants show that series extrapolations converge well down to a temperature of approximately $J/4$. At this temperature 40 percent of the total entropy of the system is still present. The heat capacity, which has a short-range order peak around $T = 2J/3$ shows hints of another rise below $T = J/4$, leading to a sharp peak in C/T. These results are reminiscent of similar results on the kagome lattice.[19–22]

Although the susceptibility data can be well fit by a Curie-Weiss law over a wide temperature range, the effective Curie-Weiss constant keeps changing with the region of fit.[23] Comparison with the experimental susceptibility data shows that the material is described by a J value of approximately $300K$.[15, 16] This J value is consistent with more recent high-field measurements.[24] There are also clear deviations below $T = J/2$, where the series extrapolation are still well convergent. These presumably arise from smaller exchange terms induced by spin-orbit couplings, such as the Dzyaloshinski-Moria interactions, which are not included in our theoretical studies.[25, 26] Similar anisotropy terms are also known to be present in the kagome antiferromagnets.[3, 22, 27]

Experimental determination of the magnetic heat capacity requires subtraction of non-magnetic contributions, which becomes increasingly uncertain with increasing temperature. Our study can not shed light on the low temperature power-laws observed in the material. At temperatures above $J/3$ which translates to $\approx 100K$ for the material, where our results are well converged, there is a clear discrepancy between the experiments and the model calculations. We show that these discrepancies can be reconciled if one assumes some additional harmonic modes below $300K$ that have not been fully eliminated by the subtraction procedure in the experimental

paper.[14]

MODEL AND SERIES EXPANSIONS

We study the nearest-neighbor spin-half Heisenberg model

$$\mathcal{H} = J \sum_{i,j} \vec{S}_i \cdot \vec{S}_j. \quad (1)$$

Here, the sum runs over each nearest-neighbor pair of spins on the hyperkagome lattice. We develop high temperature series expansions for $\ln Z$ and the uniform structure factor, which is also $T\chi$, where χ is the uniform susceptibility. These quantities are given by

$$\ln Z = \ln Tr \exp -\beta\mathcal{H}, \quad (2)$$

and

$$T\chi = \sum_{i,j} < S_i^z S_j^z > . \quad (3)$$

Here the sum runs over all the spins of the lattice, and the angular brackets denote a thermal expectation value. Details of the method for calculating the series expansions can be found in the literature.[28] Let us write

$$\frac{\ln Z}{N} = \sum_n a_n (\frac{-\beta J}{4})^n, \quad (4)$$

where N is number of sites, and,

$$\frac{T\chi}{N} = \sum_n b_n (\frac{-\beta J}{4})^n. \quad (5)$$

The coefficients a_n and b_n upto $n = 16$ are given in Table 1. Note that the series coefficients are valid for both the ferromagnetic and the antiferromagnetic couplings. The factors of $(-1)^n$ are needed for the antiferromagnetic model, where $J > 0$.

SERIES ANALYSIS AND RESULTS

We begin with an analysis of the uniform susceptibility series. To convert the susceptibility into emu/mole, we need to multiply $T\chi$ by a factor of $N_A g^2 \mu_B^2 / 4k_B = 0.0938 g^2$. At high temperatures the susceptibility can be fit to a Curie-Weiss law, whose asymptotic value for our model is $T_{cw}^{\infty} = J$. However, as discussed by Zheng et al,[23] the effective Curie-Weiss parameter obtained by fitting the inverse susceptibility versus temperature data to a straight line slowly changes with temperature. It can be defined around a temperature T as

$$T_{cw}(T) = -T - \frac{\chi}{d\chi/dT}. \quad (6)$$

n	a_n	b_n
0	$\ln 2$	0.25
1	0.000000000000D+00	0.100000000000D+01
2	0.300000000000D+01	0.200000000000D+01
3	0.000000000000D+00	0.100000000000D+01
4	-0.850000000000D+01	-0.666666666667D+00
5	0.000000000000D+00	0.134666666667D+02
6	0.465333333333D+02	0.352888888889D+02
7	0.213333333333D+01	-0.699206349206D+02
8	-0.312973809524D+03	-0.237619047619D+03
9	-0.354539682540D+02	0.745729100529D+03
10	0.235000994709D+04	0.236886624339D+04
11	0.467922116402D+03	-0.685090572391D+04
12	-0.188903919384D+05	-0.231225320571D+05
13	-0.570519634440D+04	0.656040985160D+05
14	0.158860661564D+06	0.236506418283D+06
15	0.668440957641D+05	-0.623070199393D+06
16	-0.137796110009D+07	-0.244735886559D+07

TABLE I: Series expansion coefficients for $\ln Z$ and uniform structure factor for the hyperkagome lattice Heisenberg model.

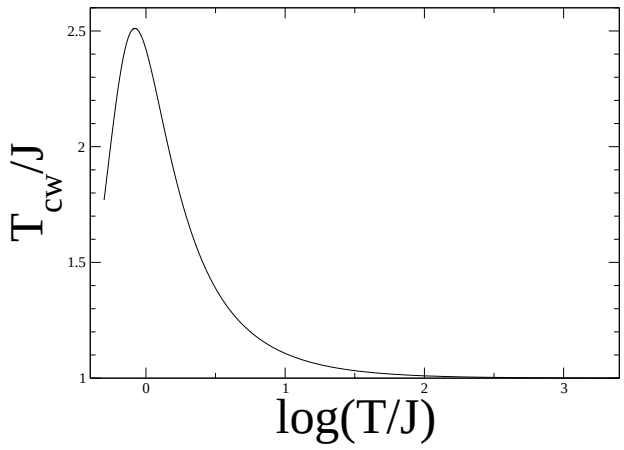

FIG. 1: Variation of the effective Curie-Weiss constant with temperature for the Heisenberg Antiferromagnet on the hyperkagome lattice.

A plot of this effective Curie-Weiss constant as a function of $\log(T/J)$ is shown in Fig. 1. We see that even at temperatures of $10J$ the effective Curie-Weiss constant is 10 percent off its asymptotic value. At temperatures around and below J the effective Curie-Weiss constant is larger than $2J$. This means that the exchange constant must be a factor of 2 or more smaller than determined by Okamoto et al.[14] Indeed, $J = 300K$ and $g = 1.99$ leads to an excellent fit of the experimental data above $T = 150K$. Since the g-factor is not independently known, a finer fit is not useful. We note that the series extrapolation converges well down to about $J/4$ and be-

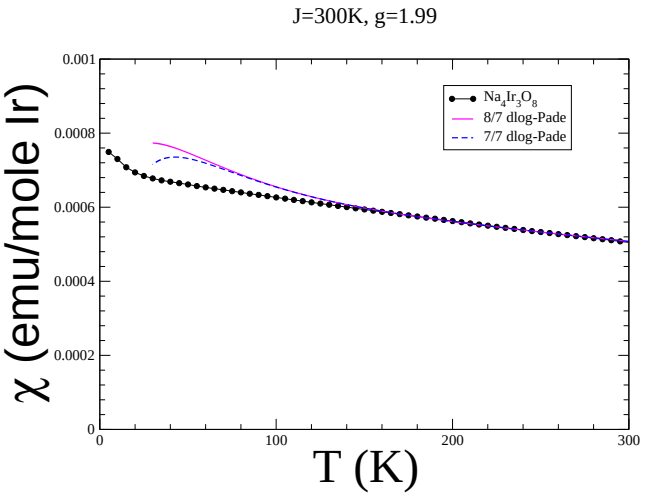

FIG. 2: A comparison of the uniform susceptibility data of $Na_4Ir_3O_8$ versus temperature with the Heisenberg model for $J = 300K$ and $g = 1.99$.

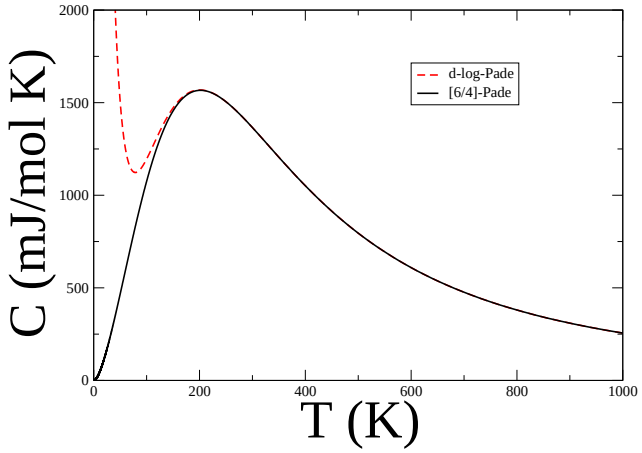

FIG. 3: The molar heat capacity for the hyperkagome lattice Heisenberg model with $J = 300K$. The [N+2/N] pade approximant ensures a T^2 behavior at low temperatures. But, it has a 40 percent missing entropy. The d-log Padé approximants imply a second peak below $T = 100K$.

low $J/2$, there are clear deviations between the experimental data and the Heisenberg model. We attribute these to spin-orbit couplings, which must be present in the material.[26] Nevertheless, the susceptibility comparison shows that the nearest-neighbor Heisenberg model is a good starting model for the material.[25] All other terms are presumably an order of magnitude smaller.

For the rest of the study, we will fix the J value to $300K$. Series extrapolations for the heat capacity are shown in Fig. 3. There is a peak associated with short-range order around a temperature of $200K$. This peak is well captured in our calculation. However, the extrapolations do not converge well below the peak. Experiments show a T^2 heat capacity at low temperatures. If we use an [N+2/N] pade to represent the heat capcity, it ensures a T^2 dependence. All such extrapolations are close to the [6/4] approximant shown. However, they can not be right. The integration of the heat capacity implies a large missing entropy of order 40 percent. A similar problem was observed in extrapolating the high temperature series for the kagome lattice Heisenberg model.[20] If instead we use d-log Padé approximants to extrapolate the series, we find that the majority of the approximants show a second rise in the heat capacity at a temperature below $100K$. However, once again the convergence breaks down as the second peak arises. The method developed by Misguich and Bernu[21] may be helpful in this regard. However, their method is based on taking advantage of our knowledge of the ground state and the low temperature properties in the extrapolation of the high temperature expansions. Since we know even less about the ground state properties of the hyperkagome lattice than we do about the kagome lattice, it is unclear how useful it would be at this stage.

A comparison of the experimental C/T data with the theory is shown in Fig. 4. We clearly cannot address the low temperature power-laws, which occur at temperatures well below our range of convergence. The discrepancy at higher temperatures can possibly be attributed to an incomplete subtraction. Experimental determination of the magnetic specific heat requires subtraction of phonon and other contributions. When the J values are comparable to room temperature, this becomes difficult as the phonon contributions are not simply power-laws. For this material, the non-magnetic $Na_4Sn_3O_8$ was used to subtract the phonons. But such a subtraction need not be accurate above some temperature. In fact, as shown in Fig. 5, the entropy obtained from the experimental data exceeds the theoretical curves above 70 K and exceeds $\ln 2$ before 200 K. In our calculations, the entropy is well converged down to about 70 K. It is also much less sensitive upto these temperatures to extrapolation methods, and to addition of other smaller exchange constants.

We believe, there can be two reasons for this discrepancy. First, the subtraction is incomplete. Indeed, if we further subtract harmonic oscillator modes with a density of states of about 0.25 moles at a single frequency of $200K$ that can bring the data into good agreement with experiments, as shown in Fig. 4. A second possibility is that the materials have additional entropy associated with itinerant degrees of freedom because they are not too far from a metal insulator transition. This issue can presumably be addressed by further experiments.

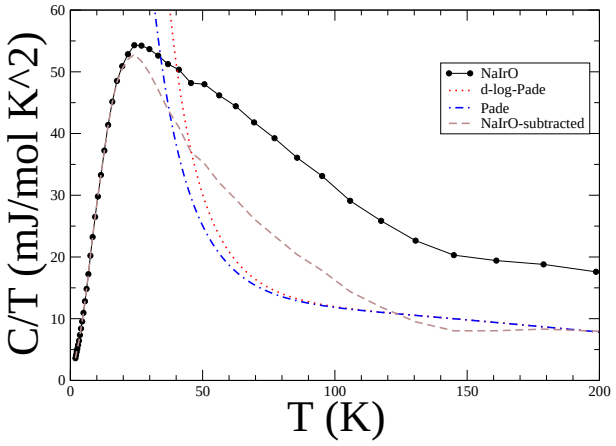

FIG. 4: Comparison of C/T between theory and experiments. The line with filled circles is the experimental data from Ref.14. The solid lines show representative behavior from series extrapolations. The dashed line is experimental data from which additional subtractions corresponding to a heat capacity of 0.25 moles of phonons at a single frequency ($200K$) has been carried out.

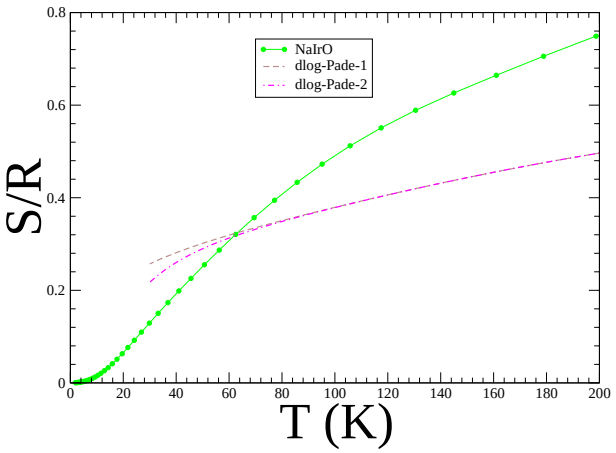

FIG. 5: The entropy function calculated for the hyperkagome lattice Heisenberg model and the experimental material.

CONCLUSIONS

In this paper, we have developed high temperature series expansions for various thermodynamic properties of the Heisenberg model on the hyperkagome lattice. These expansions enable us to compute various thermodynamic properties, such as the uniform susceptibility, the heat capacity and the entropy down to a temperature of $J/4$. Comparison with the experimental data for $Na_4Ir_3O_8$ suggests that the Heisenberg model is a good starting model for the material with an exchange constant $J \approx 300K$. However, we find that there must be additional terms also present in the material. The observed low temperature power-laws can not be addressed by our calculations. We have suggested possible resolutions to discrepancies in higher temperature heat capacity and entropy data for the material.

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